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ENVIRONMENTAL CONTROLS FOR UNDERGROUND COAL GASIFICATION: GROUND-WATER EFFECTS AND CONTROL TECHNOLOGIES

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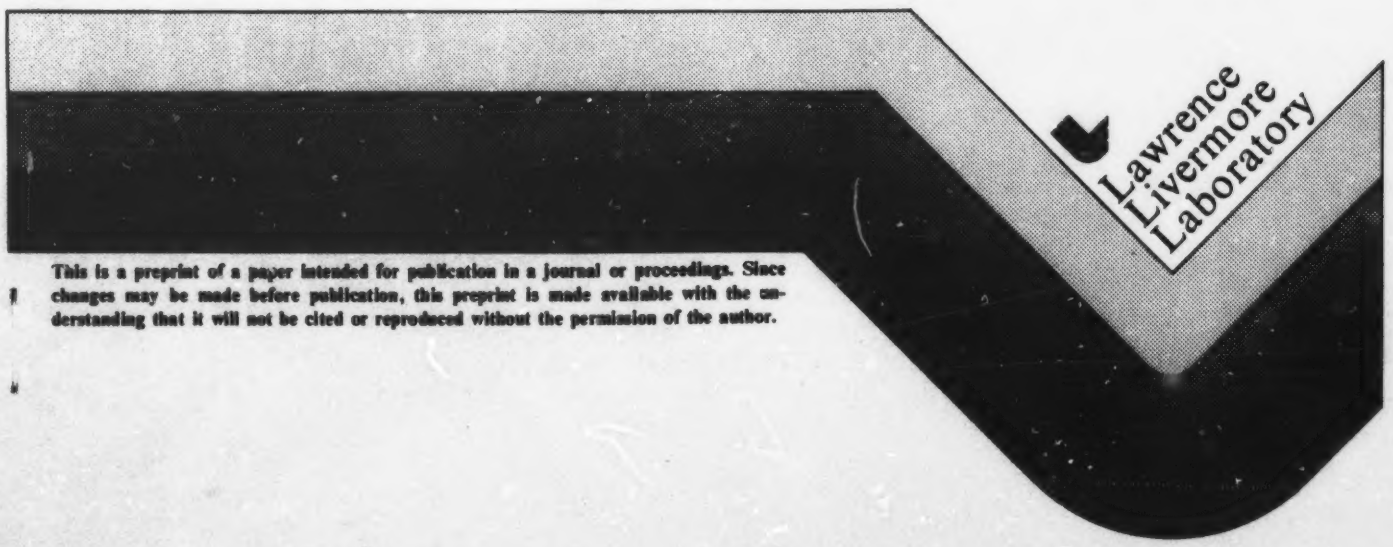

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ENVIRONMENTAL CONTROLS FOR UNDERGROUND COAL GASIFICATION: GROUND-WATER EFFECTS AND CONTROL TECHNOLOGIES *

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ABSTRACT

Underground coal gasification (UCG) promises to provide economic access to an enormous deep-coal resource. It is, therefore, of considerable importance to develop appropriate environmental controls for use in conjunction with the UCG process. The Lawrence Livermore Laboratory has conducted three UCG experiments at its Hoe Creek site in northeastern Wyoming. We are conducting environmental studies in conjunction with these UCG experiments, including an investigation of changes in local ground-water quality and subsidence effects. Ground-water monitoring and geotechnical measurements have helped to clarify the environmental significance of reaction-product contaminants that remain underground following gasification, and the implications of cavity roof collapse and aquifer interconnection. These investigations have led to the development of preliminary plans for a specific method of ground-water quality restoration utilizing activated carbon adsorption. We are also investigating unconventional technologies that may be appropriate for restoring ground water that has been contaminated as a result of UCG operations. These water treatment technologies are being explored as possible supplements to natural controls and process restrictions.

INTRODUCTION

The conversion of coal into combustible gases, as a source for synthetic fuels, promises to become an important method of coal utilization. If the conversion process is carried out with the coal in place underground - in situ gasification or Underground Coal Gasification (UCG) - there are important economic and environmental advantages. However, the exploration and assessment of this prospective technology must include a careful evaluation of potential environmental problems, together with efforts to identify effective environmental controls.

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The Lawrence Livermore Laboratory (LLL) has carried out three underground coal gasification experiments at its Hoe Creek site in northeastern Wyoming. We are conducting environmental studies in conjunction with these UCG experiments, including an investigation of changes in ground-water quality and subsidence effects. Our objectives are to characterize any ground-water or subsidence problems that may exist, and to identify appropriate environmental control technologies. The ground-water investigations are designed, primarily, to evaluate the magnitude and extent of hydraulic and chemical changes that could result from UCG. In some instances, UCG has led to cavity roof collapse and interconnection of aquifers. It is important to understand the altered hydrologic regime that will result under such circumstances, and its effect on contaminant dispersal. An understanding of both chemical and hydrologic factors is essential to the design of effective environmental controls. This paper summarizes the results of our ongoing ground-water studies at the Hoe Creek site and presents our preliminary ideas and investigations concerning possible control technologies and mitigation measures that may be applicable to ground-water quality changes that result from UCG.

BACKGROUND

In order to evaluate the environmental concerns and constraints associated with UCG, it is important to have an understanding of the process itself. Although underground coal gasification generally involves a complex series of chemical reactions, it can be simply characterized as the heating of coal in the presence of gasifying agents such as oxygen and steam. Some of the coal is burned to provide heat to drive the gasification reactions. In the simplest form of UCG, two or more process wells drilled into the coal seam are used, after the coal is ignited, to inject air or other gasifying agents and to withdraw the resulting combustible gas mixture (Figure 1). Usually, the coal's permeability must be enhanced, before gasification, along a path connecting the process wells. This may be accomplished by explosive fracturing (as was done at Hoe Creek I), reverse burn linking (Hoe Creek II), directional drilling (Hoe Creek III), hydrofracturing, or electrolinking[1]. The need to achieve this preliminary connection, reliably and economically, represents an important current challenge in UCG technology.

The possibility of ground-water contamination arises because some of the gasification reaction products remain underground in the vicinity of the gasified coal seam, which is typically an aquifer. The residual materials include coal ash, char, some coal tars, and roughly 10% of the product gases, which escape into the surrounding permeable strata. During gasification, these escaping product gases can transport small amounts of volatile contaminants up to 600 ft (200 m) or more, from the gasification cavity. Following the gasification process, the ground water, which has been prevented from entering the gasification cavity by product-gas pressure - and perhaps in part by dewatering operations - returns to the cavity, leaches inorganic materials from the ash, and dissolves some of the organic species. A contaminated ground-water plume develops and begins to move through the coal seam under the influence of the natural ground-water flow. This contaminated ground water would be of environmental significance if it found its way, in sufficient concentrations, into surface waters or into aquifers from which water is extracted for drinking or agricultural purposes. Fortunately, the concentrations of many contaminants are significantly reduced as a result of sorption by the surrounding coal.

We have monitored ground-water quality before, during and after the three Hoe Creek UCG experiments. The first experiment, performed in 1976, was comparatively small, involving gasification of only 130 tons of coal. The gasified coal seam (Felix No. 2) is 25 ft (7.6m) thick and lies at a depth of 125 ft (38 m). Phenolic materials are among the contaminants that showed large increases near the burn zone following gasification. However, the concentrations decreased rapidly with time and distance from the burn zone, as indicated in Figure 2. At distances of about 50 ft (15m) from the burn zone, concentrations have returned to baseline values. It is important to note that normal ground-water flow rates in the vicinity of Hoe Creek are quite slow - perhaps a few meters a year - and that the results exhibited in Figure 2 do not, for the most part, represent contaminant transport as a result of ground-water flow. Our laboratory measurements provide confirmation that the decrease in phenol concentrations with time is largely a result of sorption on the coal.

Some important new factors affected contaminant distribution at the sites of the more recent UCG experiments at Hoe Creek: Hoe Creek II (2300 tons of coal gasified, 1977) and Hoe Creek III (3600 tons gasified, 1979). A primary difference between these large gasification experiments and the Hoe Creek I experiment is the fact that roof collapse and the resulting gasification of an overlying coal seam (Felix No. 1) led to the interconnection of the Felix No. 2 Coal aquifer with the Felix No. 1 Coal aquifer and with an overlying sand aquifer. Roof collapse was documented by an extensive array of subsurface geotechnical instruments[2], and the nature of the permeable collapse zone was delineated in detail by post-burn coring at site II (Fig. 3)[3].

Since the Hoe Creek site is an area of natural recharge to ground water[4], we expected that water from the overlying aquifers would flow downward toward the bottom of the gasification cavity and radially outward into the Felix No. 2 Coal seam. Our measurements of ground-water contaminants and hydraulic heads as a function of time confirm the expected behavior. The radial ground-water flow is the dominant ground-water movement within 100 ft (30 m), or so, of the burn zone.

An example of contaminant transport that is believed to be a result of the accelerated ground-water motion due to aquifer interconnection is shown in Figure 4.[5] The initial high concentrations of the aromatic hydrocarbons are probably due to the rapid outward transport of these materials in the vapor phase during gasification[6]. After a decrease, perhaps due to sorption, the contaminants increase slowly as a result of ground-water transport from the more concentrated source near the gasification zone.

In Figure 5, we have plotted boron concentrations in several outer wells at various times after gasification. These results also indicate increasing concentrations in some outer wells with time, as would be expected if boron is following the natural ground-water flow and being transported outward with the moving ground water.

Another interesting feature of the contaminant distribution that resulted from the Hoe Creek II and III experiments is the fact that the highest phenol concentrations occur in the overlying Felix No. 1 Coal seam. This may be because the hydrostatic pressure within the Felix No. 1 Coal is less than that in the underlying Felix No. 2, and the cavity gas pressure is therefore more effective in driving vaporized contaminants out into the Felix No. 1 Coal seam.

ENVIRONMENTAL CONTROLS

We have begun an investigation aimed at identifying effective and economically feasible methods for controlling or mitigating ground-water quality changes caused by UCG. Our investigation has included laboratory studies to assist in evaluating the effectiveness of promising control technologies. It is important to remember that natural controls - in particular, ground-water cleansing by the surrounding coal - and UCG operational restrictions are likely to play a major role in the control and limitation of ground-water contamination.

An essential prerequisite to the development of meaningful ground-water controls is a knowledge of the applicable water quality standards. The task is complicated by the broad range of organic and inorganic products that are found in the ground water as a result of underground coal gasification (Table 1). Furthermore, the water quality standards themselves are certain to change with increased understanding of the environmental significance of coal gasification by-products. In an effort to provide a tentative basis for control technology development, we have assembled in Table 2 a listing of water quality standards as specified by the Environmental Protection Agency and the Wyoming Department of Environmental Quality. We have also listed some recommended values that may provide insight concerning future regulations.

Operational Limitations and Natural Controls

We are beginning to recognize operational characteristics of the UCG process that affect contaminant dispersal and that may be altered so as to limit contamination - often with advantageous effects on process economics. For example, the extent of contaminant deposition that results from product gas escape is, in part, dependent on cavity pressure. By keeping cavity pressure as low as possible, consistent with the need to restrict ground-water influx, it should be possible to limit the initial dispersal of gaseous contaminants, as well as the loss of valuable product gases.

Site selection criteria that preclude weak overburdens and nearby aquifers may also be beneficial - if not essential - in achieving successful process operations and effective limitations on contaminant migration.

The principal natural control that restricts ground-water contamination is the sorption by the coal surrounding the gasification zone. Our ground-water monitoring studies suggest that coal effectively sorbs many of the contaminants introduced by the UCG process.

Table 1

Ground-Water Contaminants that Increased Following Gasification at Hoe Creek Sites[5]
(Listed in approximate order of abundance.)

Organics	
Volatile Hydrocarbons:	methane, ethane, ethylene, propane
Aromatic Hydrocarbons:	benzene, touluene, C_2-C_4-benzenes, napthalenes, 3- and 4-ring aromatics, indans
Organic Acids:	phenol, alkylphenols, naphthol, alkylnaphthols
Organic Bases:	pyridine, methyl pyridine, C_2-C_4-pyridines, aniline, indolines, quinolines, isoquinolines
Saturated and Cyclic Hydrocarbons:	n-alkanes, branched alkanes, cyclic alkanes
Unsaturated Hydrocarbons:	olefins
Inorganics:	boron, flouride, bromide, chloride, lead, lithium, sulfate, manganese ammonia, sodium, sulfide, thiocyanate, iron, calcium, cyanide, barium, potassium, magnesium

It is important to consider the mechanisms that appear to be involved in various kinds of coal sorption. The coal network is generally assumed to be formed from small aromatic units held together by short cross-linkers - mainly methylene, but also including ethytene, propylene and ethers. A coal particle appears to act like a sponge. It is permeated by large and small fissures or pores (from a few tenths to 10 nm) that are more or less accessible to small molecules such as ammonia and low molecular weight hydrocarbons. Most of these volatile materials are trapped in the pores of the coal and are not actually chemically bonded[12]. In the case of small pores, the foreign molecules are able to enter them only at high temperatures and pressures, but are trapped therein upon cooling. Larger pores permit unrestricted entrance and exit of molecules, but other sorption processes may occur.

We have conducted preliminary laboratory studies on the sorption capacities of coal[13]. However, there are difficulties in duplicating the two geologic colloid systems that appear to exist underground (gas in solid phase and liquid in solid phase). The sorption of gas or vapor by coal can be described by the Langmuir isotherm[14], whereas the sorption from solution by coals is

TABLE 2

Water Quality Standards and Recommendations (Future)

Contaminant	EPA Requirements (ppm) (Future - Present)	Wyoming DEQ Requirements (ppm) (Domestic - Livestock)
Phenols	0.001 - 9.5	0.001
Carboxylic acids	? - 100	--
Organic bases	0.1 - 10	--
Polynuclear bases	10^{-5} - 10^{-3}	--
Polynuclear aromatics	10^{-6} - 10^{-3}	--
Polynuclear aromatic hydrocarbons	10^{-6} - 10^{-3}	--
Oil and Grease	1 - 15	10.0
Organic Sulfur	.1 - 10	--
Endrin	? - .0002	--
Lindane	? - .004	--
Methoxyclor	? - .1	--
Toxaphene	? - .005	--
Chlorophenoxys	.01 - .1	--
BOD	10 - 30	--
COD (State & Municipal)	30 - 500	--
CCE	0.3 - ?	
CAE	1.5 - ?	
Inorganics		
Ammonia (NH_3)	0.5 - 10	0.5 - ?
Sulfide ($S^=$)	.03 - 0.3	
Carbonate ($CO_3^=$)	--	
Cyanide (CN^-)	0.02 - 0.2	0.2 - ?
COS	--	
SCN^-	--	
Fluoride (F^-)	1.4 - 2.4	1.4 - 2.4
Cadmium (Cd)	.001 - 0.01	0.01 - 0.05
Chloride (Cl^-)	250 - ?	250 - 2,000
Aluminium (Al)	--	- 5.0
Copper (Cu)	1.0 - ?	1.0 - 0.5
Hydrogen Sulfide (H_2S)	--	0.05 - ?
Iron (Fe)	0.3 - ?	0.3 - ?
Manganese (Mn)	0.05 - ?	0.05 - ?
Sulfate ($SO_4^=$)	250 - ?	250 - 10,000
Uranium (U)	--	5.0
Zinc (Zn)	5.0 - ?	5.0
Cobalt (Co)	--	? - 1.0
Mercury (Hg)	0.0005 - 0.002	0.002 - 0.00005
Arsenic (As)	10^{-6} - 10^{-3}	0.5 - 0.2
Silver (Ag)	? - 0.05	0.05 - ?
Selenium (Sc)	.001 - 0.01	0.01 - 0.05
Lead (Pb)	.005 - 0.05	0.05 - 0.10
Boron (B)	0.5 - 2.0	.75 - 5.0
Barium (Ba)	? - 1.0	1.0
Chromium (Cr)	? - 0.05	0.05
Nitrate (as N)	? - 10.0	1.0 - 10.0
Total Dissolved Solids	500 - 5000	500 - 10,000
Color - Turbidity	1.0 NTU	
Odor	Insignificant	
Coliform Bacteria	four/100 ml	
Ph	6 - 9 s.u.	6.5 - 8.5 s.u.
Ames test for Mutagenicity	Negative	
SAR	8	8
RSC	1.25 meq/l	1.25 meq/l
Combined Total		
Radium - 226 and Radium - 228	5 pCi/l	5 pCi/l
Total Strontium	8 pCi/l	8 pCi/l
Gross alpha particle radioactivity	15 pCi/l	15 pCi/l
Tritium	20,000 PCi/l	--

References: (7), (8), (9), (10), (11)

best described by the Freundlich isotherm[14]. The main difference is that sorption of gases and vapors onto solids is primarily a function of temperature and pressure, while the sorption capacity of solids from solution depends upon the pH of the solution, concentration of the solute, contact time, and competition adsorption reactions between species. Although we have measured, in the laboratory, the sorption of many ground-water contaminants in solution on granular coal at pH 7, we have not yet been able to predict absolute sorption characteristics under field conditions[13]. In many geologic colloid systems, such as coal, adsorption is expected to play a significant role. It is also important to consider the rate at which desorption takes place. Desorption depends upon many factors including changing solution characteristics and the type of adsorption process that occurred initially. Furthermore, thermodynamic equations demonstrate that if adsorption occurs from a solution, the surface excess (number of adsorbed layers) increases with increasing concentration of the adsorbate in solution. If the adsorption occurs from the gaseous phase, the surface excess increases with increasing partial pressure of the adsorbate[14].

The outstanding ability of activated carbon to adsorb various organics and inorganics from solution is well known[12]. Our recent laboratory experiments have shown that bituminous coal is the best source of activated carbon for the removal of organics that are present in the ground water affected by UCG operations. The first step in the production of activated carbon is the formation of a char from the source material (carbonization). This is accomplished by heating the source material (usually in the absence of air) to a temperature that is sufficiently high to dry and volatilize substances in the carbon[12]. To increase the surface area, the carbon is then activated by the action of oxidizing agents such as steam, air or CO_2 at elevated temperatures. This results in the development of an extremely porous structure and an extensive internal surface area with much greater adsorption capacity. The adsorptive properties of the finished product depend heavily on the oxidizing gas, its concentration and the temperature during oxidation.

Since activated char is formed during the gasification process[15], it may be possible to provide increased adsorption capabilities in the surrounding coal by promoting the formation of this uniquely effective adsorbent. The experiment at Hoe Creek I used explosive fracturing to enhance permeability prior to gasification. This may have provided additional surface area for activation, thereby leading to the formation of more highly activated coal surfaces. In fact, contaminant levels have decreased most conspicuously at Hoe Creek I (compared to Hoe Creek II) but there are many other factors that could equally well account for the observed differences. Nevertheless, the possibility of providing increased adsorption through special activation procedures deserves further consideration.

Supplementary Control Measures

If a combination of process controls and sorption by the surrounding coal proves inadequate for effective control of ground-water quality, then supplementary measures will need to be implemented. There are three approaches that can be explored: (1) minimizing contaminant transport (2) external removal of contaminants or (3) converting contaminants to innocuous

substances. We have begun to evaluate possible control technologies that could be both effective and economically feasible. Figure 6 suggests the steps involved in assessing potential cleanup techniques. Much of the required information is already available to us as a result of our ground-water sampling investigations.

Our current investigations include a consideration of conventional water treatment methods which involve pumping the contaminated water to the surface. If pumping can be initiated soon enough after gasification - before the normal ground-water flow has extended the region of contamination - it may be possible to create a flow pattern that prevents further contaminant dispersal. Preliminary estimates suggest that reasonable pumping capabilities (less than 100 gpm) may be adequate. Although this approach seems straightforward, we are uncertain of its ultimate effectiveness - primarily because slow desorption of residual contaminants may prolong the effective lifetime of the contaminant source. Experiments designed to provide an estimate of this effect are under consideration. Of course, chemical additives might hasten the desorption process. In any case, it must be remembered that a combination of several techniques may be most effective.

We are presently involved in process feasibility and engineering design studies to determine system effectiveness and cost economics for ground-water treatment in connection with large scale UCG operations. We are evaluating a combination of conventional wastewater treatment techniques, as well as some possible in situ methods (Fig. 6). In situ treatment might involve oxidation, ozonation, biological, chemical or containment methods. We have contracted with both National Technical Services (NTS), Corvallis, Oregon, and NUS Corporation of Pittsburgh, PA to develop design criteria and cost estimates for conventional treatment methods that may be applicable to UCG operations. We have conducted laboratory jar tests and bench scale studies (involving Hoe Creek water samples) in collaboration with NTS. The techniques studied included activated carbon adsorption, wet-air oxidation, aeration and ozonation. Our results suggest that certain powdered activated carbons in appropriate amounts are effective in removing problem organics. As indicated previously, activated carbon prepared from bituminous coal was found to be the most effective adsorbent. Total organic carbon concentrations were reduced to <0.1 ppm in Hoe Creek water samples whose initial TOC values were ~75 ppm. Concentrated synthetic solutions made up in Hoe Creek water using typical contaminants and having TOC concentrations of about 280 ppm were lowered to <10 ppm in these bench scale studies. Aeration and ozonation were found to be effective preliminary treatments and might be incorporated into the system design, since air and oxygen are both available on site. A preliminary design for surface treatment of UCG-contaminated ground water is indicated in Figure 7. Note that carbon regeneration is involved. It is of interest that some treatment methods which have been considered too costly to be used in conjunction with surface gasification may be cost effective when applied under the conditions that prevail during UCG operations. The disposal of treated water may involve reinjection or, possibly, repeated use in continuing gasification process operations.

In summary, we emphasize that underground coal gasification is a promising technology that may provide an important contribution to our needs for clean, easily transportable fuels. To insure that UCG development is not unnecessarily delayed, it is imperative that significant environmental effects be anticipated. Our studies show that a broad range of organic and inorganic contaminants are introduced into the ground water, but that most of these are substantially reduced and circumscribed as a result of sorption by the surrounding coal. In recognition of the possibility that coal cleansing and operational limitations may be judged inadequate, we are evaluating the cost and effectiveness of various ground-water restoration methodologies.

ACKNOWLEDGMENTS

The extensive program of ground-water quality sampling and analysis at Hoe Creek - which forms the essential basis for our efforts to develop effective control technologies - is carried out under the supervision of Francis T. Wang and Daniel H. Stuermer. We are also grateful to Harold Ganow for the geotechnical measurements and post-burn coring investigations, and to Randolph Stone for measurements and interpretations of the hydrologic regime at Hoe Creek.

Robert E. Thompson, National Technical Services, Corvallis, Oregon, has been of considerable assistance in our efforts to evaluate current water treatment techniques and explore unconventional technologies that may be appropriate for restoring ground water that has been contaminated by UCG operations.

Finally, it is a pleasure to acknowledge the continued support and assistance provided by Douglas R. Stephens, Project Director for LLL's In Situ Coal Program.

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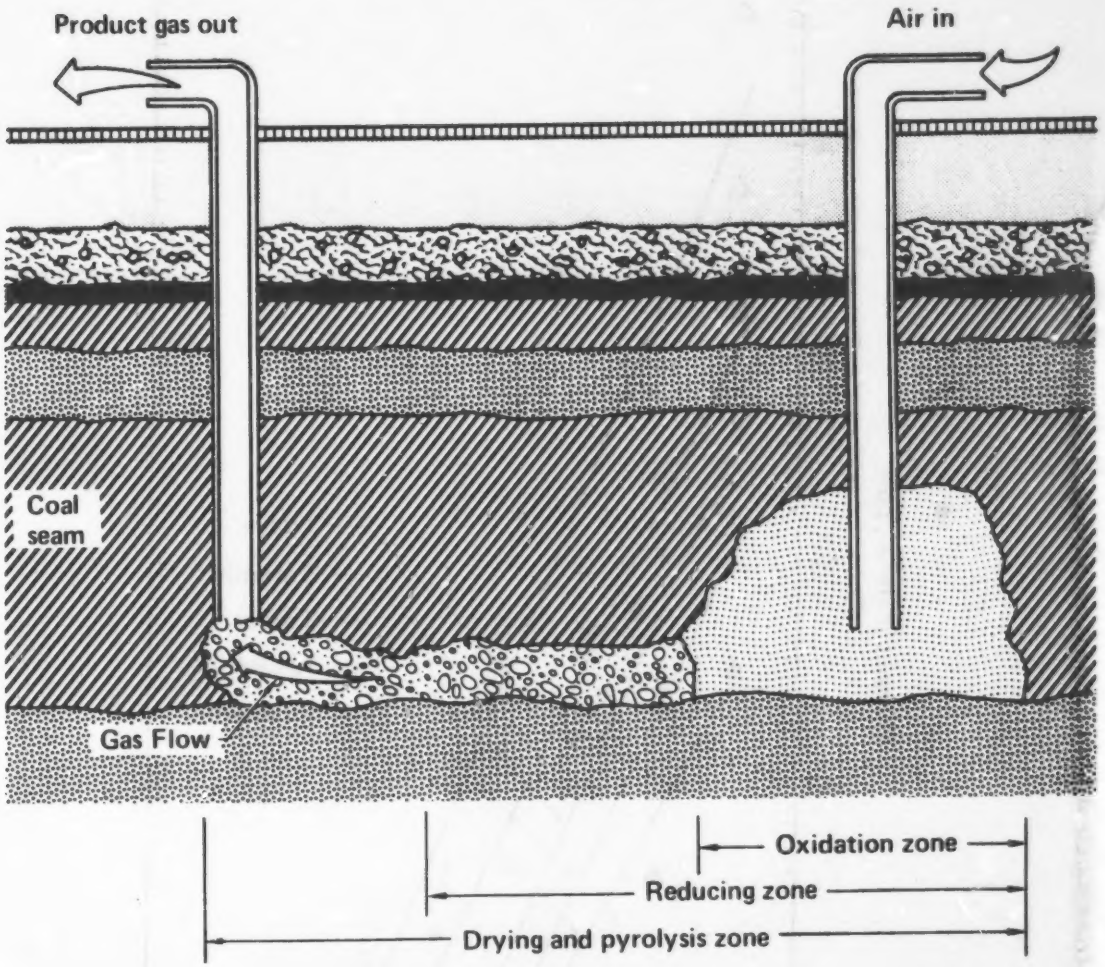

Figure 1. Schematic representation of the underground coal gasification process. The region ahead of the oxidation zone has been modified before gasification to provide a path of increased permeability that will permit adequate gas flow.

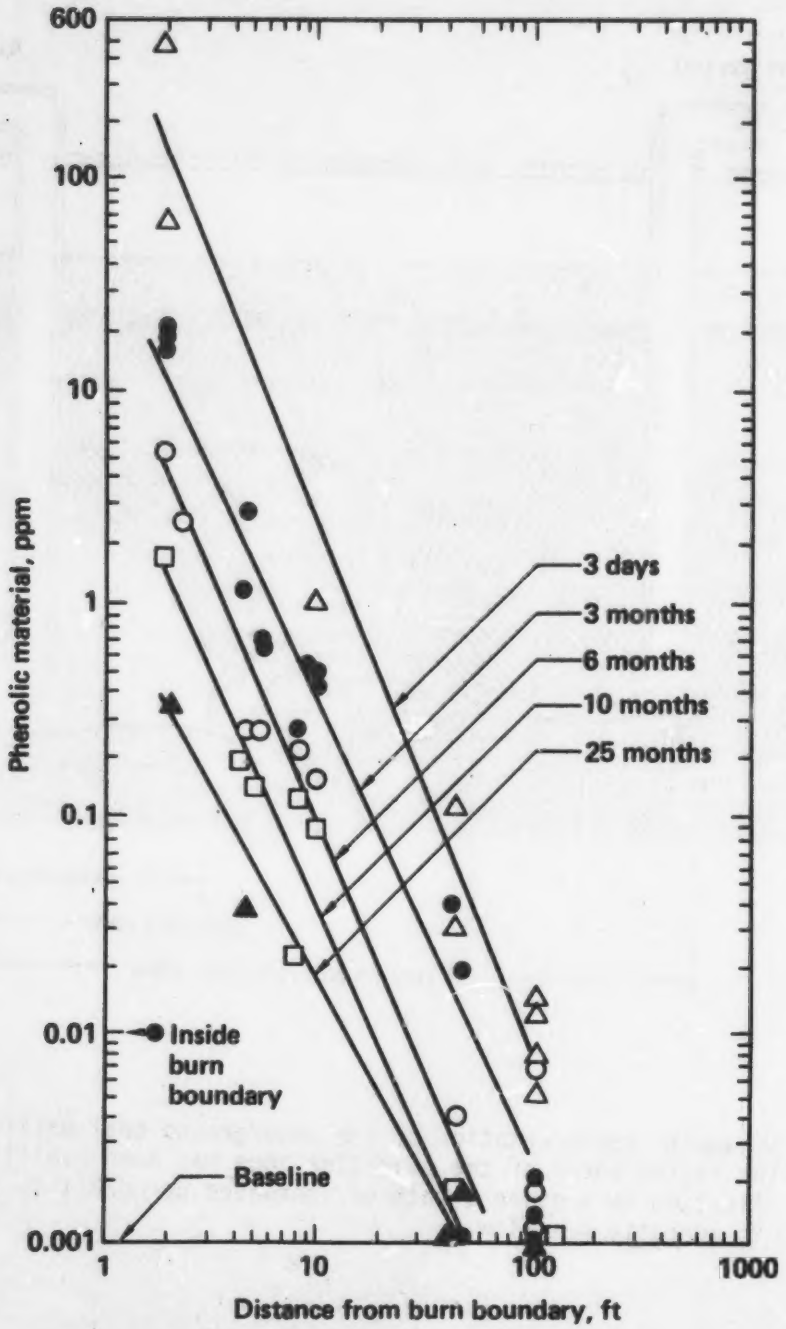

Figure 2. Concentrations of phenolic materials in the ground water as a function of distance from the nearest burn boundary of the Hoe Creek I gasification experiment. Times are measured from the end of gasification. The sampling wells are completed in the gasified coal seam, and located in various directions from the gasification zone.

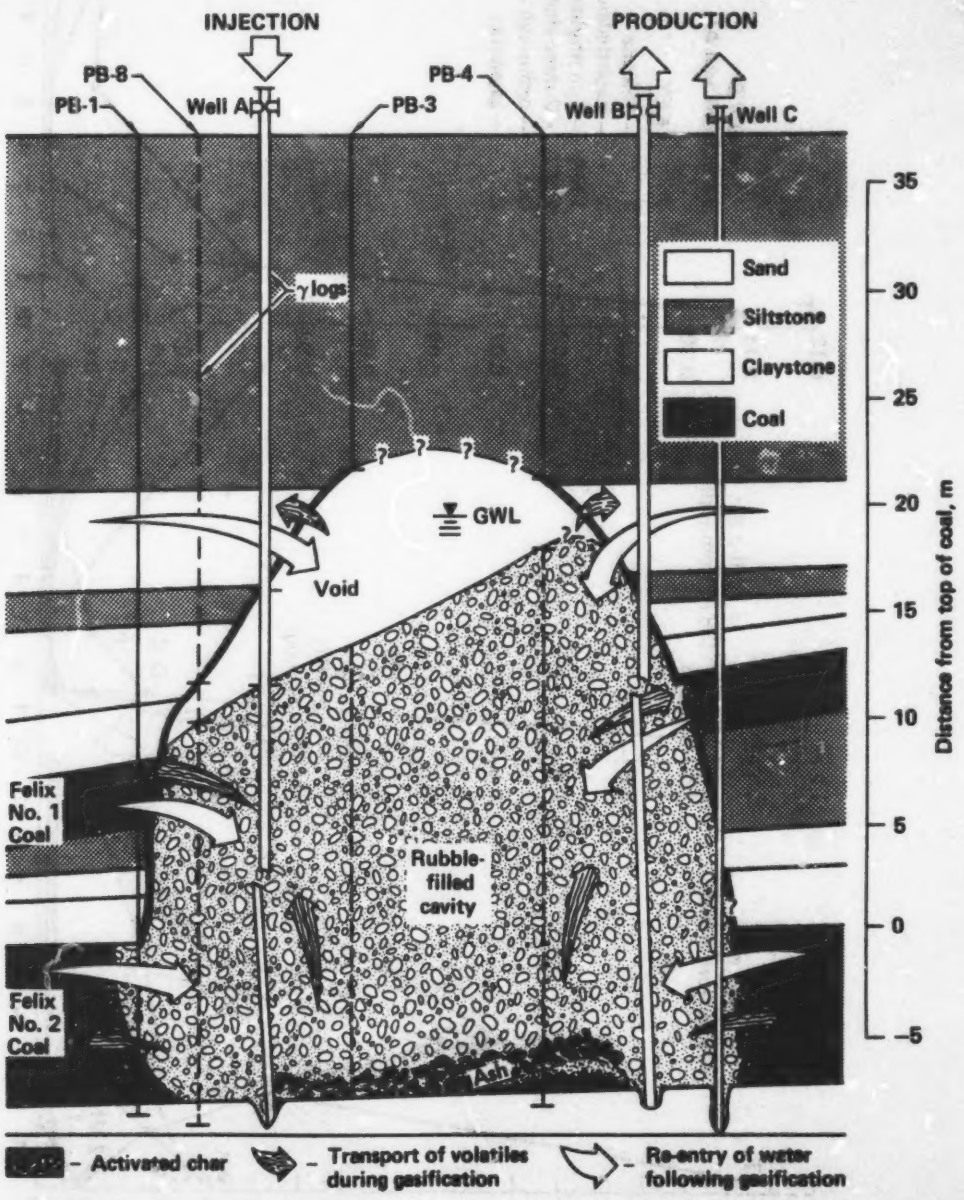

Figure 3. Inferred shape of the Hoe Creek II gasification cavity on the basis of post-burn coring and logging investigations. Roof collapse and gasification of the overlying Felix No. 1 Coal contributed to the vertical extension of the cavity.

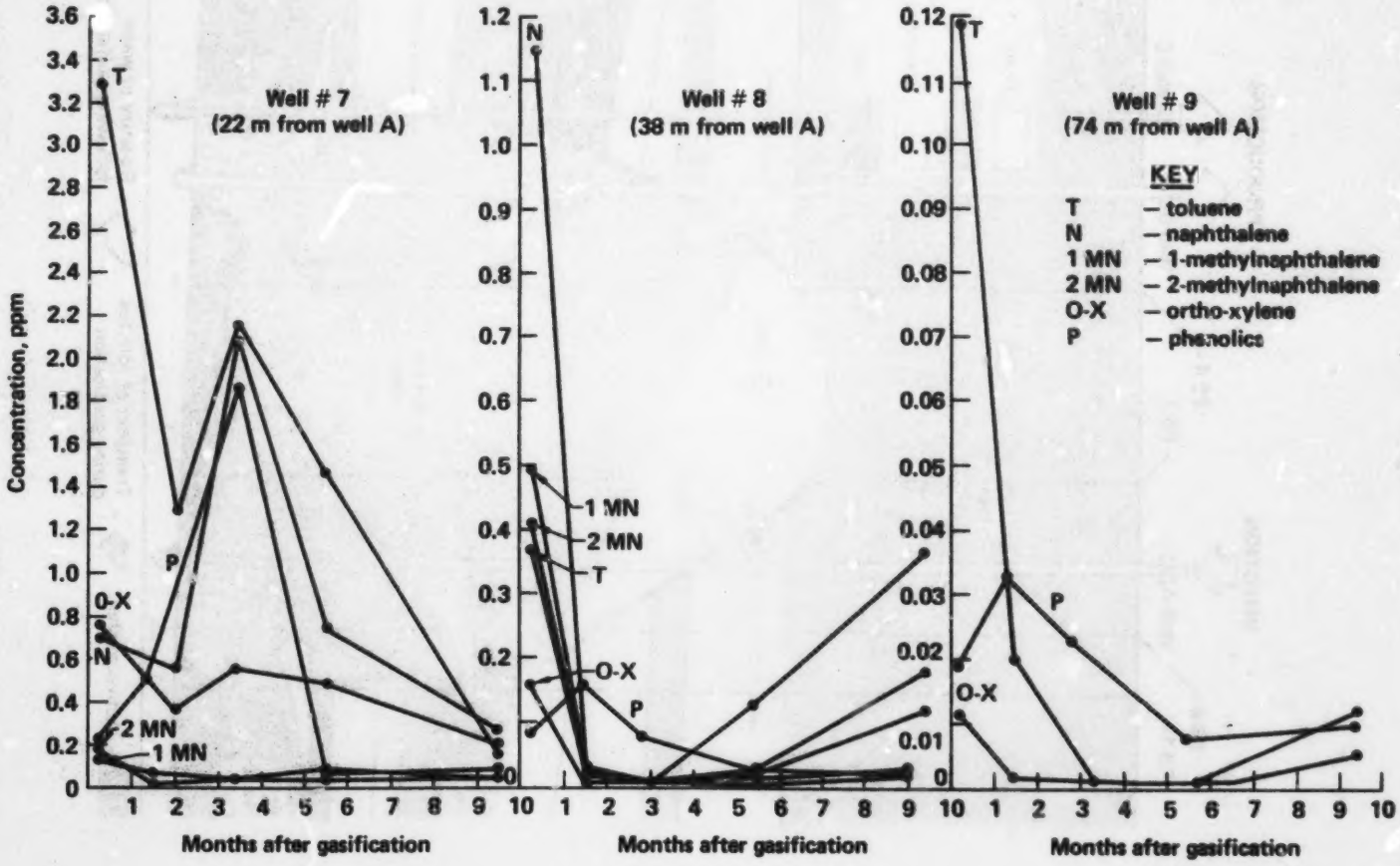

Figure 4. Changes in the concentrations of aromatic hydrocarbons and phenols as a function of time in three wells near the Hoe Creek II gasification experiment.

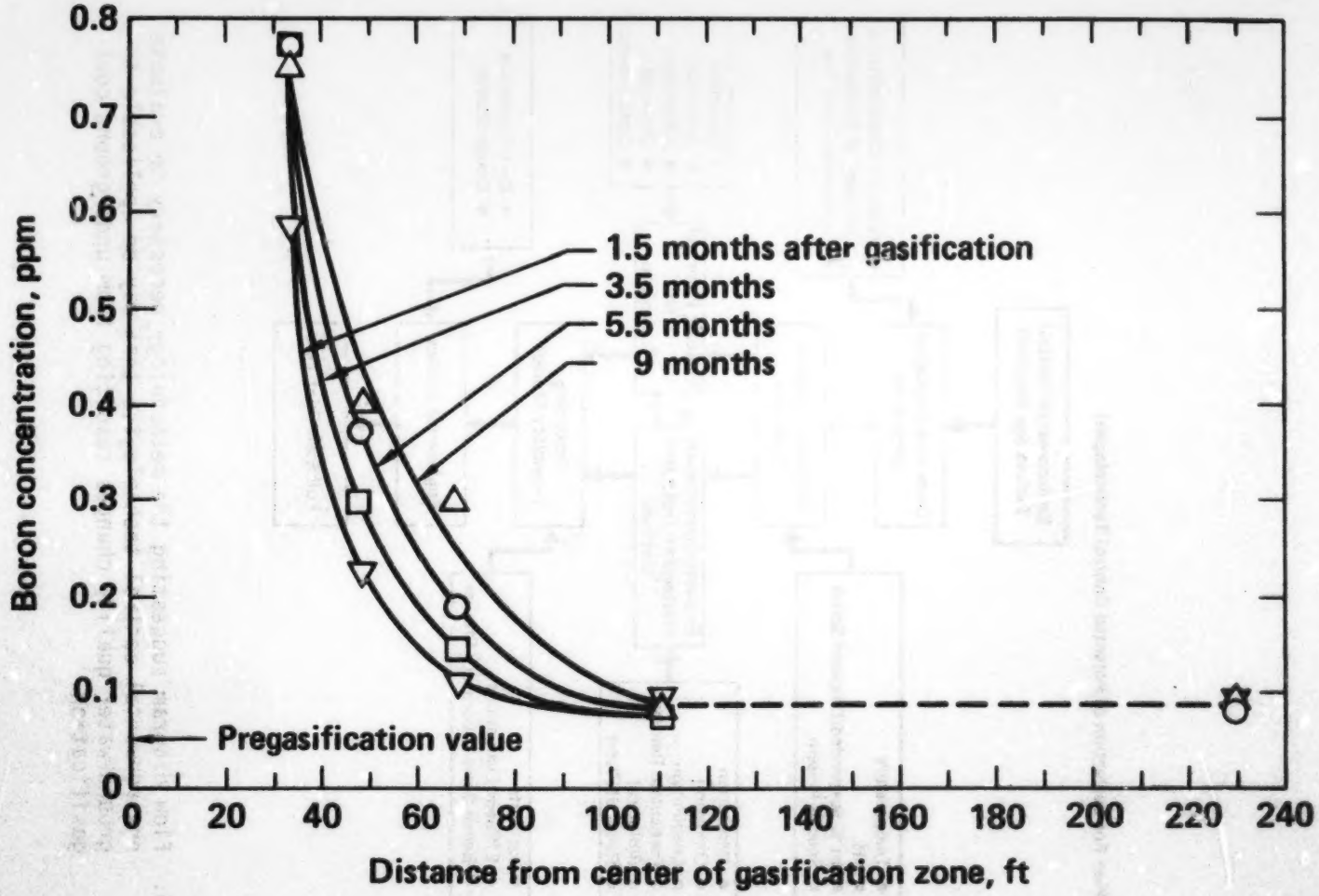

Figure 5. Changes in the boron concentrations measured in ground-water samples obtained near the Hoe Creek II gasification site.

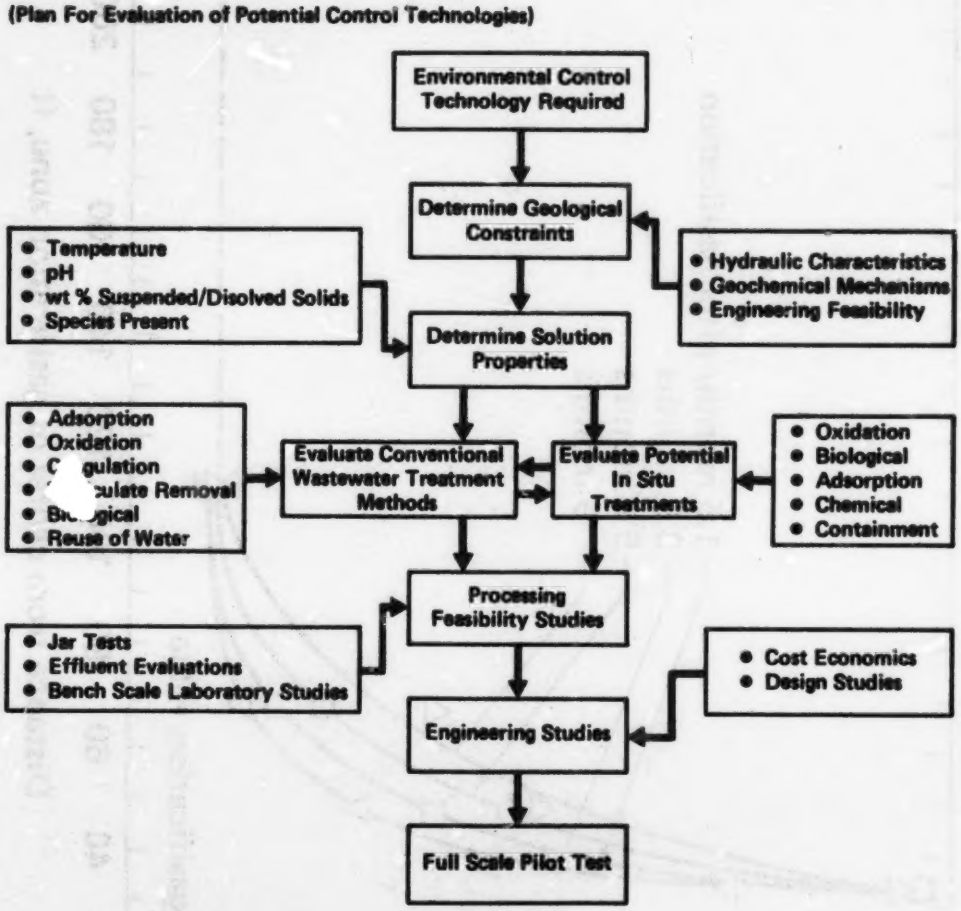

Figure 6. Flow diagram suggesting the methodology necessary to evaluate environmental control technologies that may be applicable to ground-water quality changes resulting from underground coal gasification.

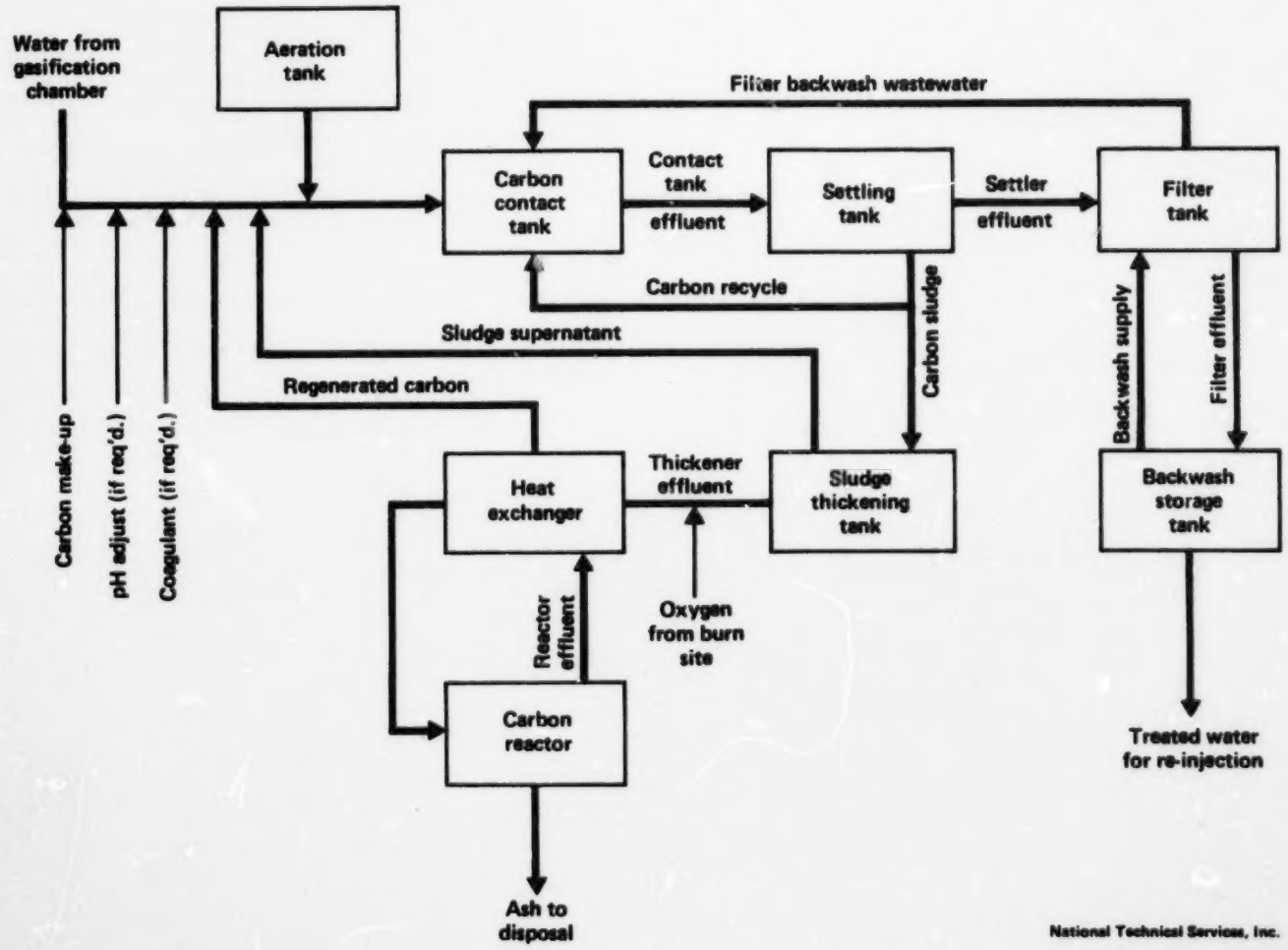

Figure 7. Schematic representation of a possible wastewater treatment facility for surface cleanup of ground waters affected by UCG operations. This system, designed by National Technical Services, Corvallis, Oregon, utilizes activated carbon adsorption.